

**10 yr trends in
atmospheric Hg at
Svalbard**

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Ten years trends in atmospheric mercury concentrations, meteorological effects and climate variables at Zeppelin, Ny-Ålesund

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Abstract

Results from ten years of gaseous elemental mercury (GEM) measurements at Zepelin Station, Ny-Ålesund, Svalbard, show no overall annual trend between 2000 and 2009. Seasonal trend analysis showed significantly decreasing trends in January, February, March and June and significantly increasing trends in May and July through December. Results showed that atmospheric mercury depletion events (AMDEs) were equally distributed between April and May with only a few having been observed in March and June. A negative correlation between AMDEs and temperature is reported and supports earlier observations that AMDEs tend to occur at low temperatures. Lower concentrations of GEM were seen at lower temperatures below a threshold of 0°C. The occurrence of AMDEs and wind direction were well correlated with the lowest GEM measured when the wind direction was from the Arctic Ocean region. Wind speed was found to not correlate with AMDEs, but the lowest GEM concentrations were observed at low wind speeds between 4 and 11 ms⁻¹. AMDEs and relative humidity did not correlate well, but the lowest GEM levels appeared when the relative humidity was between 80 and 90%. Diurnal variation was observed especially during the month March and is likely due to daytime snow surface emission induced by solar radiation. Relationships between GEM concentration and the Northern Hemisphere climate indices were investigated to assess if these climate parameters might reflect different atmospheric conditions that enhance or reduce spring AMDE activity. No consistent pattern was observed.

1 Introduction

Mercury (Hg) is a major pollutant in the Arctic environment, and there is evidence of increasing concentrations in some marine ecosystems over time (AMAP, 2011). The presence of Hg in traditional foods contributes to increasing concern about food safety for Arctic indigenous people. Atmospheric Hg is recognised as one of the main sources

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to the environment and particular attention has been paid to understand the importance of atmospheric mercury depletion events (AMDE) as a means of delivering Hg to polar ecosystems (AMAP, 2011).

Hg in the atmosphere exists in a variety of forms as gaseous elemental mercury (GEM or Hg⁰), gaseous oxidized mercury (GOM) and particle bound mercury (PBM). The residence time of GEM in air is approximately 6 months to 2 yr and is sufficient to allow for homogeneous mixing within the hemisphere of origin (Schroeder et al., 1995). The residence time of GOM and PBM are much shorter and they can in general be deposited within hours to weeks (Schroeder and Munthe, 1998).

In 1995 it was discovered that during spring unexpectedly low concentrations of GEM occurred in the Arctic air (Schroeder et al., 1998). These phenomena were termed atmospheric mercury depletion events (AMDEs) (Schroeder et al., 1998), and were later confirmed to occur throughout polar regions: at Barrow, Alaska (Lindberg et al., 2002b), Ny-Ålesund, Svalbard (Berg et al., 2003), Kuujjuarapik, Quebec (Poissant, 2003), Station Nord, Greenland (Skov et al., 2004), Amderma, Russia (Steffen et al., 2005), Andøya (Berg et al., 2008b) and Antarctica (Ebinghaus et al., 2002; Pfaffhuber et al., 2012). During AMDEs a series of photochemically initiated reactions involving halogens, convert GEM to more reactive species that subsequently associate to particles in the air and/or deposit to the polar environment (Steffen et al., 2008) and references therein). The halide species (e.g. Cl, Br, ClO, BrO) are probably derived from heterogeneous gas/solution reactions at the interface of hygroscopic sea salt aerosols, which may be initiated in the oceans (Steffen et al., 2008).

Worldwide atmospheric mercury measurement up to the early 2000s suggested that GEM levels increased from the late 1970s to a peak in the 1980s and decreased to a plateau around 1996 to 2001 (Slernr et al., 2003). At Mace Head, air masses arriving after having traversed the North Atlantic Ocean, have shown a decrease in the total gaseous mercury (TGM) concentrations of $-0.028 \pm 0.01 \text{ ng m}^{-3} \text{ yr}^{-1}$ for the period 1996–2009 (Ebinghaus et al., 2011). Long term atmospheric mercury monitoring data in polar regions of GEM are rare. However, GEM has been measured at Alert,

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Canada since 1995. A trend analysis of the data have shown a decreasing trend of $-0.0086 \text{ ngm}^{-3} \text{ yr}^{-1}$ for the 1995–2007 period (Cole and Steffen, 2010). Trend analyses have earlier also been carried out on parts of the Hg data from Zeppelin: Berg et al. (2004) reported no trend for GEM concentrations for the period 1994 to 2002.

5 When an extended time-series (1995–2005) was considered Berg et al. (2008a) again reported no significant trend at Zeppelin. In both previous studies from Zeppelin, manually collected data for 1994–1999 were included. In the first study annual averages were used. In the last study, annual, winter, spring, summer and autumn means were used.

10 Intensive measurement campaigns designed to understand atmospheric mercury cycling can provide considerable information which apply to a snapshot in time and care should be taken when scaling-up such measurements to apply for a whole year (Pfaffhuber et al., 2012). Long term programs can yield critical information to better understand the processes involved in the cycling of Hg in the polar atmosphere. Long-term data of GEM is also important for understanding the effects of climate warming on the mercury cycle. Additionally, long term data is needed to see possible effects on AMDEs and long term deposition of Hg to the polar regions from changes in the global anthropogenic Hg emissions (Steffen et al., 2008).

20 In the present manuscript, trend analysis of GEM has been carried out to see if there was a change in the concentrations from 2000 to 2009. The changes in trends have also been studied for each month. Additionally, the relationships between AMDEs and different meteorological, UV radiation and climate parameters have been examined.

2 Methods

2.1 Study site

25 Ny-Ålesund is a small settlement near sea-level on the western coast of Spitsbergen (Fig. 1). The air sampling is performed at the research station on the nearby Zeppelin

Mountain (78° 54' N, 11° 52' E; 474 m a.s.l.). The station is located on a mountain ridge, with steep hills to the north and south, and higher mountain peaks (1000–1500) to the west and east. The mountain station was situated as such to minimize the influence of local pollution from the Ny-Ålesund area.

2.2 Sampling

Measurements of GEM were carried out using a Tekran gas phase mercury analyzer (model Tekran 2537A, Tekran Inc.) (Berg et al., 2003). The basic principle of operation is that the sample air stream goes through gold cartridges where GEM is collected. GEM is then thermally desorbed and detected by cold vapour atomic fluorescence spectroscopy (CV-AFS). The Tekran 2537 was set up with a heated sampling line and an extra Teflon filter (2 µm, 47 mm) at the inlet of the sampling line. A soda lime trap was also installed in line before the instrument filter. Due to the presence of two filters and the soda lime trap the measurements represent GEM and very little or no GOM (Steffen et al., 2002). The soda lime trap was changed weekly. The sample inlet was located 3 m above ground. Auto calibrations were carried out daily using the internal calibration source and were verified by manual injections regularly. A sampling flow rate of 1.5 L min⁻¹ and a sampling time of 5 min were used. The 5 min means have then been averaged to hourly or daily means. The detection limit of the Tekran 2537 A is < 0.1 ng m⁻³, and the precision was found to be within 5% (Aspmo et al., 2005). The Tekran 2537A provide volumes at 0 °C and 760 mm Hg (Tekran Inc., 1998).

Meteorological measurements on an hourly basis from the Zeppelin station (temperature, wind direction, wind speed, relative humidity and atmospheric pressure) were provided from NILU. Data for precipitation has been provided from the Norwegian Meteorological Institutes measurements in Ny-Ålesund.

Measurements of UV erythema (McKinley and Diffey, 1987) hourly dose rates were obtained from the ground-based Ultraviolet Radiometer (GUV, Biospherical Instruments Inc.) on top of the Sverdrup Station in the Ny-Ålesund settlement at 25 m a.s.l.). Applying the libRadtran simulation package (Mayer and Kylling, 2005), the

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erythral UV measurements were transformed to the corresponding UVB and CRSO3 hourly dose rates on top of the Zeppelin mountain station (474 m a.s.l.) assuming the same ozone and cloud conditions. The CRSO3 dose rates were computed using the wavelength-dependent ozone absorption cross-sections (Molina and Molina, 1986) as
5 action spectrum. The erythemally-effective UV dose rates were converted to UVB dose rates because UVB radiation seems related to Hg measurements (Lindberg et al., 2002b).

Monthly sea ice area index and maps for the Northern Hemisphere was obtained from the (Fetterer et al., 2012).

10 An atmospheric depletion episode is defined in this study as GEM concentrations below 1.0 ngm^{-3} .

2.3 Statistics

The trend analysis was carried out using the Seasonal Kendall test for trend (a variant of the Mann–Kendall test) and the related Sen’s slope estimator (Gilbert, 1987). The Seasonal Kendall test for trend used hourly data points and treated each point within
15 the month as a replicate point for the month. The Sens calculation of slope was then performed to give an estimate of the linear slope, showing trends of different magnitudes and directions in all the months. If seasonal trends were homogenous, the results were used to determine an overall trend for the period. Otherwise, less precise but ho-
20 mogenous seasonal trends and the overall trend were calculated using monthly median GEM.

GEM concentrations during and after the active AMDE period (February to June) were also compared to meteorological parameters at the Zeppelin station. For this analysis, hourly concentrations in a given month were binned according to a corre-
25 sponding hourly meteorological parameter such as temperature or wind speed. Within each bin, summary statistics were calculated for the hourly GEM values as long as the bin contained at least 20 hourly GEM values.

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Finally, correlation coefficients for monthly median GEM concentrations and monthly climate indices were calculated using the Statistica software program. Monthly values of standard Northern Hemisphere teleconnection indices were obtained from the National Weather Service Climate Prediction Center (Fetterer et al., 2012).

3 Results and discussion

The arithmetic mean and median for the dataset were $1.54 \pm 0.23 \text{ ngm}^{-3}$ and 1.60 ngm^{-3} , respectively. This is in agreement with the mean average concentrations for the Northern Hemisphere, $1.5\text{--}1.7 \text{ ngm}^{-3}$ proposed by Slemr et al. (2003), but significantly higher than in the Southern Hemisphere as evidenced by the annual GEM mean concentration of 0.93 ngm^{-3} at the Norwegian station Troll in Antarctica from 2007 to 2011 (Pfaffhuber et al., 2012).

The entire Zeppelin station data set for the period 2000–2009 is presented as a monthly box and whisker plot in Fig. 2. The monthly median concentrations are quite high during late fall, winter and early spring. This is the time of the year when transport from mid latitude source regions dominates. In April and May the concentrations are at their absolute lowest due to the AMDEs. During summer the concentrations are increasing again due to the reemission of previously deposited GEM. In late summer the concentrations start decreasing again and by fall the levels are approximately at the global background for the Northern Hemisphere.

3.1 Trends in atmospheric GEM

The time series of GEM daily average levels including the trend line over the period of 2000 to 2009 is presented in Fig. 3. The annual concentration changes calculated using the Sen's slope estimate (based on the seasonal Kendall test for trend) found no significant annual trend for the GEM data from Zeppelin between 2000 and 2009, either at the 90 or 95 % confidence level (Fig. 3).

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The results of the seasonal Kendall test (using hourly data points) showed, however, significant trends in every single month with a 95 % confidence level. The Sens calculation of slope was then performed to give an estimate of the linear slope for each month. This shows trends of different magnitudes and directions for all months of the data set and is presented in Fig. 4. A negative slope indicates a decreasing trend in the concentration of GEM during that particular month over the period 2000–2009. Similarly, a positive slope represents an increasing trend for a given month. As seen in Fig. 4, most months show increasing trends at the Zeppelin station. February showed the largest decreasing trend of $-0.0149 \pm 0.0017 \text{ ng m}^{-3} \text{ yr}^{-1}$. January, March and June also revealed decreasing trends in GEM concentration. All other months showed significant increasing trends with the highest seen in May ($0.0287 \pm 0.0016 \text{ ng m}^{-3} \text{ yr}^{-1}$).

The lack of an annual trend in the median GEM concentration for this time period is in agreement with earlier trend analyses for Hg at this location which found no significant changes in the Hg level between 1994 and 2002 (Berg et al., 2004) or the period of 1994 to 2005 (Berg et al., 2008a). In both previous studies manually collected data were included for 1994 to 1999. In the first study the non-parametric Mann–Kendall Test and the Sen’s slope estimator were used on annual weighted means. In the second study, the same test was used on annual, winter, spring, summer and autumn weighted means. The reason the manually collected data was not included in the present trend analysis is that the quality of the measurements was not as good as for the new automatic data. This lack of a significant decreasing or increasing trend is different from what has been shown for other sampling sites. A weak decreasing trend of $-0.0086 \pm 0.0014 \text{ ng m}^{-3} \text{ yr}^{-1}$ ($-0.56 \% \text{ yr}^{-1}$) was reported for the Arctic station Alert for the period of 1995 to 2007 (Cole and Steffen, 2010). At Mace Head, Ireland, a greater decreasing trend of $-0.028 \pm 0.01 \text{ ng m}^{-3} \text{ yr}^{-1}$ ($1.6\text{--}2 \% \text{ yr}^{-1}$) was reported for the period of 1996 to 2009 (Ebinghaus et al., 2011). There could be several reasons for the reported weak or insignificant trends in the Arctic atmospheric GEM when compared to Ireland and these have been discussed in detail in Cole et al. (2012). Most of the available time-series cover the period from 1995 to 2005 (Berg et al., 2004, 2008a; Temme

et al., 2007; Cole and Steffen, 2010) during which total global anthropogenic emissions of Hg have not markedly changed, although there have been significant changes in emissions from particular source regions during this period (Pacyna et al., 2010). Hg emissions from East Asia increased by 50 % from 1990 to 2005 whereas emissions from Europe and North America have declined over the same period, resulting in smaller emissions trends overall compared to the 1970s and 1980s (Pacyna et al., 2009). Although the emission source region amounts have shifted, GEM concentrations at Zeppelin station are impacted by European emissions and not directly transport from South-East Asia (Hirdman et al., 2009). The decrease in GEM in January, February and March at Zeppelin can be a reflection of the reductions in European emissions since this is the time of the year when direct transport from Europe is the main source (Hirdman et al., 2009). Asian contributions to surface GEM at Arctic sites were shown in the GRAHM model and were similar to or lower than contributions to surface GEM in North America (Durnford et al., 2010). Emission estimates do not exist for the last part of the presented measurement period at Zeppelin (2006–2009). The long-term trends observed in the Arctic can also be controlled by trends in transport/circulation patterns that influence the air masses that reach the Arctic (Kahl et al., 1999). The lack of a decreasing trend could be due to enhanced evasion of GEM from the ocean due to decreases in ice cover (Fetterer et al., 2012). During a summer expedition in the North Atlantic on board the German research vessel Polarstern, GEM showed a homogeneous distribution over the open North-Atlantic (median $1.53 \pm 0.12 \text{ ng m}^{-3}$), in contrast to higher concentrations of GEM observed over the sea ice (median $1.82 \pm 0.24 \text{ ng m}^{-3}$) (Aspmo et al., 2006). It was hypothesized that this resulted from either (re-)emission of mercury contained in snow and sea ice surfaces that had been previously deposited during AMDEs in the spring or evasion from the ocean due to increased reduction potential at high latitudes during the summer. During another expedition in the Arctic Ocean, enhanced DGM concentrations were measured below the ice which may indicate that the sea ice acted as a barrier for evasion of mercury from the Arctic Ocean to the atmosphere (Andersson et al., 2008). As well, elevated DGM concentrations

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were observed in water that might have originated from river discharge. (Andersson et al., 2008). During the same expedition, higher concentrations of GEM were also seen along the sea ice route when compared to the marine boundary layer over the ice-free Arctic (Andersson et al., 2008; Sommar et al., 2010). The authors proposed that should an abrupt warming occur in the future, the current cycling of Hg in the Arctic will be largely perturbed and sea-air interactions will be an important source of mercury to the atmosphere. Andersson et al. (2008) calculated a hypothetical average Hg⁰ evasion of 60 ng m⁻² day⁻¹ during the Beringia 2005 expedition with a maximum as high as 2300 ng m⁻² day⁻¹. St. Louis et al. (2007) calculated an Hg⁰ flux of 130 ± 30 ng m⁻² day⁻¹ at ice break-up. In-turn, Outridge et al. (2008) predicted an eight-fold increase in summer Hg⁰ flux from the Arctic Ocean for the future between 60° N and 80–90° N.

The seasonal trend analysis for Alert (1995–2007) showed decreasing trends in all months except for May which showed a positive trend (Cole and Steffen, 2010). At Zeppelin only January–March and June showed decreasing trends in the present study covering 2000–2009. The rest of the months showed positive trends. A recent study covering both stations and the same period (2000–2009), showed some similarities and some differences (Cole et al., 2012). From January to August the two stations showed similar trends. January–March and July showed decreasing trends for both the stations, but the trends were in general stronger for Alert. April and May showed positive trends for both stations, but April was stronger for Alert. The increasing trend seen at Zeppelin in April and May could be due to less AMDEs and higher GEMs and is discussed in the next sections. September to December showed in general positive trends for Zeppelin and negative trends for Alert. The increasing trend seen at Zeppelin in fall could be due to reemission from the Arctic Ocean where the sea ice is at a minimum during this time of year and have been decreasing. This is discussed in detail in Cole et al. (2012).

3.2 AMDEs: seasonal and annual variations

Episodic depletions in GEM concentrations are seen during the months following polar sunrise, which occurs on 19 February at this latitude, and have been discussed in the literature (Berg et al., 2003; Aspmo et al., 2005; Steen et al., 2011). During AMDEs concentrations can drop from 1.8 ng m^{-3} to well below 0.1 ng m^{-3} (Aspmo et al., 2005). The AMDEs normally did not start until at least one month after polar sunrise (late March). Figure 5a shows the monthly percent distribution of AMDEs for 2000 to 2009. AMDEs are distributed quite evenly between April and May as 44 and 42 %, respectively (Fig. 5a). Fewer AMDEs occur in March and June where they are reported 6 and 8 % of the time, respectively. There is no pattern of change between when the frequency of depletion events occurred. For example, 2001, 2003 and 2007 were years when April was the predominant month for AMDEs and in 2000, 2004 and 2006 most AMDEs occurred in May. In 2002 and 2005 there was an equal distribution of AMDEs between the two months. No AMDEs have been reported earlier than in March. For the years where AMDEs were predominately in April, the average temperatures for April and May were -12.3 and -5.7 °C, respectively (Fig. 5b). The average temperatures for the years when AMDEs were equally distributed between the two months were -10 °C for April and -5.5 °C for May. For the years when AMDEs mostly occurred in May the average temperatures were -5.2 in April and -4.5 °C in May. Lower average temperatures seem to correlate with AMDEs in April. AMDEs and their temperature correlation is discussed in detail in the next section.

The percent of hourly measurements that was classified as AMDEs in March, April, May and June for the period 2000–2009 are presented in Fig. 6. The most AMDEs were seen in 2000 and decreased to a minimum in 2006. The last three years of the dataset, 2007–2009, show an increase in the number of AMDEs. Recent changes in the climate and sea ice cover in the Arctic Sea is likely to have strong effects on the cycling of mercury in Polar Regions. While the local fjord ice is likely not an important source for AMDEs at Zeppelin, it can be used as an example of changes in the sea

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that are occurring on a local scale. Ny-Ålesund is located at the fjord Kongsfjorden. The fjord was in general ice-covered in the past. However from 2006 to 2008 there was no permanent ice on the Kongsfjorden. Cottier et al. (2007) showed that, during the winter of 2005/2006, periods of sustained along-shelf winds generated upwelling and cross-shelf exchange causing extensive flooding of the coastal waters with warm Atlantic water from the West Spitsbergen Current. The winter temperature of the West Spitsbergen Shelf reverted to that of typical fall and interrupted the normal cycle of sea ice formation in the region (Cottier et al., 2007). The warm sea water from 2005/2006 was not fully replaced with cold water in 2006/2007 and 2007/2008 preventing sea ice formation on the local fjords during these winters. By studying the ice mass cover in the Arctic, large changes can be seen for most of Arctic as well as for Svalbard during the same time (Fig. 7a, b). The halide species necessary for the AMDEs are presumably derived from the sea ice (Steffen et al., 2008).

3.3 Correlation of AMDEs with meteorological and climate parameters

GEM concentrations were calculated as a function of various parameters including temperature, wind direction, wind speed, relative humidity, time of day, simulated UVB dose rate and CRO3 dose rate. The results are shown in Fig. 8a–g. For each of those parameters the data were first divided by month then the distribution of GEM values was plotted as a function of the value of that parameter. Each vertical line indicates the dividing line between each bin (e.g. temperature is binned into 5 degree increments, such as -25 to -20°C , -20 to -15°C etc.). For all the data in that bin, the distribution of GEM values has been shown using a box plot, indicating median, 25th and 75th percentiles (box), and 10th and 90th percentiles (whiskers).

A correlation was found between GEM concentrations and air temperature during the spring months such that when the temperature was below -5°C the GEM levels decreased (Fig. 8a). In general the concentrations decreased with decreasing temperature for all the months and temperature bins between -30 and 0°C . March data had few low concentrations of GEM which is expected given the timing of AMDEs as

discussed above. Lower concentrations of GEM were seen in May than in April for most of the temperature bins below 0 °C. In June, all the low GEMs occurred when the temperature were between -5 and -10 °C, which were the lowest temperatures recorded for that month. These relationships support earlier studies on the temperature dependence of AMDEs with the formation of brines, the chemistry of bromine, as well as reactivity of bromine radicals with GEM. The freezing of sea water, which causes brine to be pushed to the surface of the newly forming sea ice, is caused by cold temperatures (less than -13 °C) and is likely involved in providing a saline surface on which halogen activation can occur (Simpson et al., 2007). Adams et al. (2002) showed that the conversion of bromide ions in aerosol or snow to Br₂ may be enhanced at lower temperatures. This may either be due to the increasing concentration of halides in the reactive liquid layer of the snow pack (Koop et al., 2000) or acidification of sea salt aerosol (Sander et al., 2006). Theoretical calculations of the reaction rate in the reactions that oxidize GEM to GOM have predicted an inverse temperature dependence at the low temperatures (less than -13 °C) seen in the Arctic during spring (Goodsite et al., 2004). A large fraction of the AMDEs seen at Ny-Ålesund are suspected to be the results of long-range transport from areas with lower temperatures (less than -13 °C) where the chemistry could be initiated, e.g. the Arctic Ocean (Gauchard et al., 2005; Steen et al., 2011). Although the local temperature is not always sufficiently low to initiate the depletion chemistry, the temperatures may have been low enough at the origin of the advected air masses (Berg et al., 2008b). The highest GEM concentration levels reported at the Zeppelin site were seen on the warmest days in March (0 to -5 °C) (Fig. 8a). This was likely re-emission of previously deposited mercury as earlier shown by Steen et al. (2009) where from February to mid-June 2008, GEM fluxes above a snow covered surface were measured in Ny-Ålesund using a GEM flux gradient method. In that study, the GEM emission from the surface snow to the air was most pronounced from 24 March until 12 May suggesting that deposition of RGM and PHg during AMDEs is the main precursor for the emitted GEM. Cole and Steffen (2010) suggested that when the snow melts and sunlight penetrates further into the snow, the

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photo reduction of oxidized Hg to GEM is increased. In addition, the authors suggested that releases of Hg from biological activity may also be temperature dependent.

GEM concentration levels and wind direction were also correlated on short time scales (Fig. 7b). The lowest GEM values in the data set ($< 0.5 \text{ ngm}^{-3}$) were seen in April when the wind was blowing from north/north west. Air masses from this direction arrive from the Canadian Arctic or Arctic Ocean. Westerly winds (from Greenland) in April gave GEM values somewhat higher ($0.5\text{--}1.0 \text{ ngm}^{-3}$). In May the same relative pattern as for April was found but the concentrations were not as low. North-easterly winds (from Siberia) in March, April and May result in GEM concentrations just below 1.0 ngm^{-3} and, in June; the lowest GEM levels were from the north (Central Arctic). During the AMDE period, the highest GEM concentrations were reported in March when the winds came from south (Fig. 8b) and, in April and May, the highest concentrations were also seen with southerly winds. The source and sink regions for the data between 2000 and 2008 have recently been thoroughly examined using a Lagrangian particle dispersion model (Hirdman et al., 2009). The conclusions from this study were as follows: low GEM concentrations in April/May were strongly associated with low-level transport of air masses across the Arctic Ocean, providing evidence for the influence of AMDEs at Zeppelin station. The average GEM concentrations in air masses with strong Arctic Ocean surface contact were 33 % lower than in air masses having little or no such contact, showing a net removal of GEM from the air during April and May. Apparently, not all mercury deposited during AMDEs can be reemitted as GEM during the same season (Hirdman et al., 2009). This supports the view that mercury accumulates in the snow pack during spring (Lindberg et al., 2002a; Brooks et al., 2006). A fraction of the mercury deposited during AMDEs can be immediately re-emitted, as suggested by Lalonde et al. (2002). In April and May, high measured GEM concentrations were associated with transport from mid-latitude source regions. In July and August, the highest measured GEM concentrations were associated with strong surface contact over the Arctic Ocean which could be explained by re-emission of previously deposited mercury when snow melts in summer, and/or by the evasion of GEM from the ocean (which is

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supersaturated with dissolved gaseous mercury) when the sea ice breaks and direct contact between ocean and atmosphere is facilitated. A statistical analysis for ozone (Hirdman et al., 2009) revealed that in spring low ozone concentrations were found in air masses having ice contact, indicating a common occurrence of ODEs and AMDEs.

5 However, in summer ozone continues to be low in air masses with surface contact in the Arctic Ocean, as expected. In winter, transport of anthropogenic GEM emissions from mid-latitude source regions (especially Europe) leads to the highest observed GEM concentrations at Zeppelin. During winter the lowest GEM concentrations were associated with downward transport of air masses from the free troposphere (Hirdman et al., 2009). Using a trajectory climatology model Eneroth et al. (2007) found that the lowest concentrations of GEM at Zeppelin (2000–2001) arrived from Siberia (north-east), the Barents Sea (east) and the Norwegian Sea (south west) in April/May, and from the Canadian Arctic (north-west) and the Arctic Ocean (north) area in May/June. It was suggested the one month lag between the two source regions could either be
10 due to the position of the marginal ice zone or temperature differences between the north-western and north-eastern air masses.

Wind speed does not have a high correlation with GEM concentration (8c). However, the lowest GEM values were seen with a wind speed between 4 and 11 ms⁻¹ in April/May. Low wind speeds are commonly reported during AMDEs and can be the result of a stable marine boundary layer (Steffen et al., 2008). The highest concentrations were seen in March when the winds were between 10 and 12 ms⁻¹ (Fig. 7c). The correlation of AMDEs with wind speed was reported to be weaker than with temperature at both Alert and Amderma (Cole and Steffen, 2010). A slight decrease in GEM concentrations was seen at around 5 ms⁻¹ for all months at Alert but no relationship was seen for Amderma.
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Relative humidity did not appear to have strong correlations with GEM concentrations (Fig. 8d). The lowest GEM values were seen in April when the relative humidity was between 80 and 90 %, which is just above the April average for the station. In May, the lowest concentrations were seen when the relative humidity was 60–70 %. The highest

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GEM concentrations were seen in April when the relative humidity was in the range 40–50 % (Fig. 8d).

Slight diurnal variations in the GEM concentrations were observed (Fig. 8e). This is in contrast to GEM measurements at NILUs Troll station in Antarctica where no diurnal pattern was observed (Pfaffhuber et al., 2012). The main difference between the two stations is that Troll is located on snow free bedrock whereas the area surrounding Zeppelin is snow covered large part of the year. The diurnal pattern at Zeppelin is likely caused by daytime snow surface emissions induced by solar radiation. The solar maximum is in hour 12.

UV radiation does not have a strong correlation with GEM concentrations (Fig. 8f). The AMDEs (GEM below 1.0 ng m^{-3}) seem to be most likely to occur when the UVB dose rates are in the range $150\text{--}250 \text{ mW m}^{-2}$ (Fig. 8f) and the CRSO3 dose rates are between 40 and $70 \times 10^{-18} \text{ mW m}^{-2}$ (Fig. 8g). On an interannual basis, years in which the average UVB dose rate for March was high ($> 13 \text{ mW m}^{-2}$) had high monthly median GEM levels ($> 1.8 \text{ ng m}^{-3}$) compared to $1.5\text{--}1.7 \text{ ng m}^{-3}$ in years when the average UVB dose rate was $> 10 \text{ mW m}^{-2}$. This may be tied to increased snow surface emissions since the higher levels are above hemispheric background. In contrast, median GEM concentrations for April were inversely correlated with the average UVB dose rate on an interannual basis, which suggests that AMDE chemistry is the source of the anti-correlation. This is supported by the hourly data (Fig. 8f) and the fact that the range of median April GEM concentrations was $1.35\text{--}1.62 \text{ ng m}^{-3}$, below hemispheric background. The competition between these two effects (surface emission and oxidation) may explain the poor correlations overall.

In addition to the local meteorological parameters, large scale transport patterns and the quantity and quality of sea ice may have an effect on the GEM levels measured at Zeppelin. Altered atmospheric fields imply the possibilities of altered transport of Hg into and out of the Arctic (AMAP, 2011). The North Atlantic Oscillation Index (NOA) is the most important index for climate variability in the Northern Hemisphere. For example, during the past decade, the Arctic atmospheric pressure pattern has been

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characterized by anomalously high sea-level pressure on the North American side of the Arctic and low pressure on the Eurasian side (Overland et al., 2008), which has then supported more southerly wind. The correlation of monthly median GEM concentrations with the Northern Hemisphere climate indices was calculated for every month, with the resulting correlation coefficients listed in Table 1. This analysis was done to see if these climate parameters reflect different atmospheric conditions that enhance or reduce spring AMDE activity from year to year. As well, this was done to assess whether enhanced transport during the non-AMDE seasons is associated with higher GEM concentrations. From Table 1 it is clear that there are no consistent patterns where GEM concentrations correlate with these indices. A few cases show relationships between the parameters and climate indices that are statistically significant ($p < 0.05$) for a single month. This may be an artefact of the number of relationships examined; given a single data set with a large number of variables, some will randomly have significant statistical relationships with each other. No correlation was seen for the NAO and GEM at Zeppelin. In contrast, at Alert a significant, negative correlation was seen between AMDEs and both the NAO and the Polar/Eurasia Teleconnection, indicating that AMDEs were more intense when the circumpolar vortex was strong (Cole and Steffen, 2010). In March there was a negative relationship between Greenland Sea Ice, the Northern Hemispheric ice, the Kara and Barents Sea ice, the Arctic Sea ice and GEM at Zeppelin: years with more sea ice in these areas had more AMDEs and lower GEM, perhaps due to more surfaces for bromine activation. In September there were also negative relationships between Northern Hemispheric ice, Arctic Ocean ice and GEM: years with more ice in these areas were correlated with the lowest GEM concentrations, perhaps because of less evasion from the ocean to the atmosphere. No correlation was found between Greenland Sea ice and GEM at this time of the year, probably due to the fact that the Greenland Sea ice has almost disappeared by September.

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4 Conclusions

Gaseous elemental mercury measurements (GEM) at the Zeppelin station have shown no annual increasing or decreasing trend for the period from 2000 to 2009. This result is in agreement with earlier studies made from the same station, but is different from observations from Alert, Canada (also in the High Arctic) and Mace Head, Ireland (temperate, coastal region) where significant annual decreases in GEM concentrations have been seen over the same time period. The differences observed between the three stations may reflect changes in the anthropogenic emissions of GEM. Total global anthropogenic emissions of GEM have not significantly changed during this period, but there have been significant changes reported from particular regions. Emissions from Europe and North America have decreased but emissions from China have increased. The lack of an annual trend at Zeppelin could reflect changes in the transport and/or circulation patterns that influence the air masses that reach this area of the Arctic. However, there were no strong correlations with NAO, AO with the GEM concentration data from Zeppelin and therefore we conclude that this is not the likely reason for the lack of trend. Another suggestion for the lack of annual trends could be increased reemission from an Arctic Ocean where the ice cover is decreasing due to climate warming.

The Zeppelin GEM data showed decreasing trends in January, February, March and June but the remainder of the months showed increasing trends. The trends for Zeppelin and Alert were similar from January to August. over the same time period. For the remainder of the year the two stations showed opposing trends. The decrease in GEM in the period from January to March was attributed to reductions in European emissions since this is the time of the year when direct transport from Europe has been shown to be the main source. The increasing trend in April and May might be due to less AMDEs and a resulting higher GEM. In fall the increasing trends could be due to reemission from the Arctic Ocean where the sea ice is at a minimum and decreasing.

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The occurrence of AMDEs is equally distributed between April and May at Zeppelin Station, but a few have been recorded in March and June and never recorded earlier than one month following sunrise which is 19 February. The percentage of AMDEs in March, April, May, June compared to the total number of measurements have varied over the years.

AMDES were correlated with several meteorological variables factors such as temperature, wind direction, wind-speed and relative humidity to assess any significant relationships. Lower concentrations of GEM were seen at lower temperatures below a threshold of 0 °C. These relationships support earlier studies on the temperature dependence of AMDEs with the formation of brines, the chemistry of bromine, as well as reactivity of bromine radicals with GEM. The lowest GEM values were observed when wind speeds were between 4 and 11 ms⁻¹ in April and May. Low wind speeds are commonly reported during AMDEs and can be found when the marine boundary layer is stable and not well mixed. The lowest GEM values were observed in April when the relative humidity was between 80 and 90 %. Slight diurnal variations in the GEM concentrations were observed, and this is likely caused by daytime snow surface emissions that can be induced by solar radiation and increases in temperature. Ultra Violet Radiation, total sea ice concentration and different climate indices did not correlate with monthly GEM concentrations overall, although some statistically significant relationships exist in certain months.

The results in this study have shown that GEM at Zeppelin, Svalbard behaves differently from other stations in the high Arctic and in Europe both on short and long time scales. Ten years of GEM data have been too little to compare with different climate indices since those are in general operated on thirty years scale. Continued monitoring of GEM are therefor highly recommended both at Zeppelin, and other locations. Long term time series of GEM and other atmospheric mercury species are also important to better understand the processes involved in the cycling of Hg in the polar atmosphere and to see possible effects on AMDEs and long term deposition of Hg to the polar regions from changes in the global anthropogenic Hg emissions.

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Table 1. Correlations of median monthly GEM with climate indices, and regional sea ice. Coefficients in bold are significant at the $p < 0.05$ level. AO = Arctic Oscillation; NAO = North Atlantic Oscillation; EA = East Atlantic Pattern; PNA = Pacific/North American Pattern; EA/WR = East Atlantic/West Russia Pattern; SCA = Scandinavia Pattern; POL = Polar/Eurasia Pattern. Sea ice regions are defined by NSIDC and shown in Fig. 1.

	AO	NAO	EA	PNA	EA/WR	SCA WR	POL	Northern Hemisphere ice	Greenland Sea ice	Kara and Barents Seas ice	Arctic Ocean ice
Jan	-0.50	-0.70	0.72	0.93	-0.44	0.63	0.32	0.77	0.79	0.33	0.23
Feb	0.48	0.62	0.31	0.39	0.22	-0.34	-0.38	0.31	-0.10	0.15	0.66
Mar	0.57	0.65	-0.25	0.01	0.33	0.64	0.10	-0.61	-0.78	-0.47	-0.32
Apr	-0.36	0.11	0.29	0.47	-0.14	0.34	-0.08	-0.32	-0.57	0.38	-0.08
May	0.47	0.35	0.51	0.12	0.25	0.02	0.35	-0.10	0.09	0.10	-0.39
Jun	-0.08	0.07	-0.07	0.49	0.14	0.40	-0.82	-0.17	-0.22	0.12	-0.25
Jul	0.52	0.10	0.74	0.34	0.29	0.21	0.41	-0.11	-0.37	0.17	-0.04
Aug	-0.11	-0.05	0.07	0.04	-0.45	0.21	-0.08	-0.45	-0.16	-0.05	-0.43
Sep	-0.66	-0.12	-0.06	0.29	-0.09	0.31	0.24	-0.76	0.51	-0.12	-0.75
Oct	0.38	0.38	-0.11	0.32	-0.22	-0.13	0.07	-0.58	0.45	-0.27	-0.55
Nov	-0.41	-0.06	0.10	0.19	-0.10	-0.02	-0.51	-0.53	0.29	-0.35	-0.48
Dec	-0.19	-0.41	0.66	0.04	-0.00	0.32	-0.34	-0.30	0.20	-0.24	-0.30

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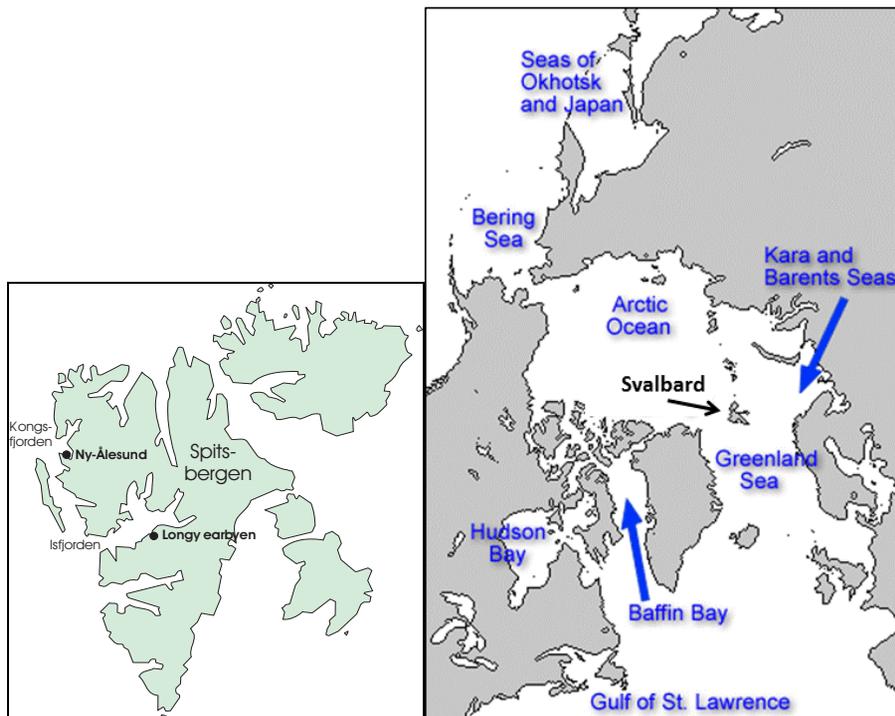


Fig. 1. Map showing Ny-Ålesund, Svalbard (left) and the sea ice regions (right).

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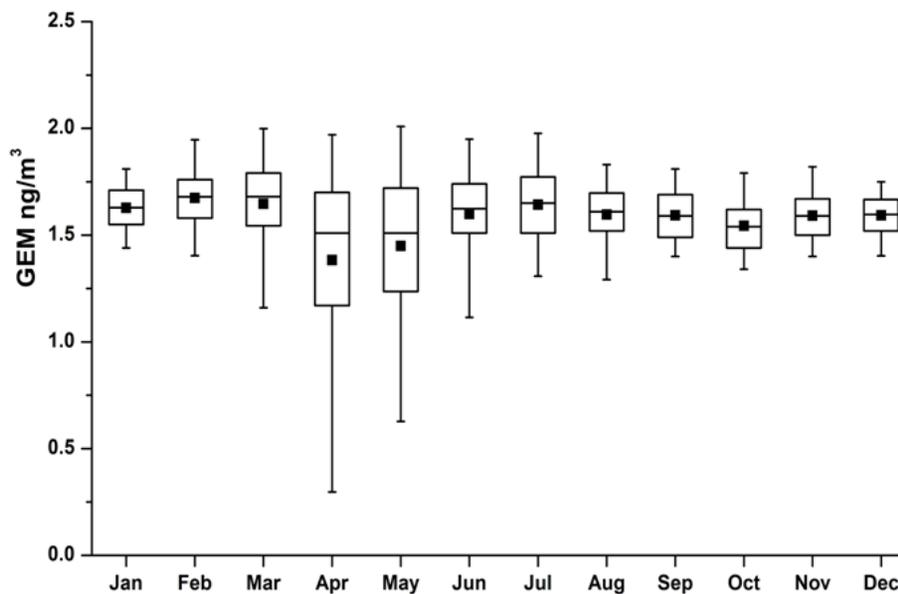


Fig. 2. Box and whisker plot presenting the concentration distribution of GEM measurements at Zeppelin. The middle line in the box shows the median concentrations, the box indicates the range between the 25th and the 75th percentile. Whiskers above and below the box indicate the 90th and 10th percentile respectively. The filled square shows the arithmetic mean.

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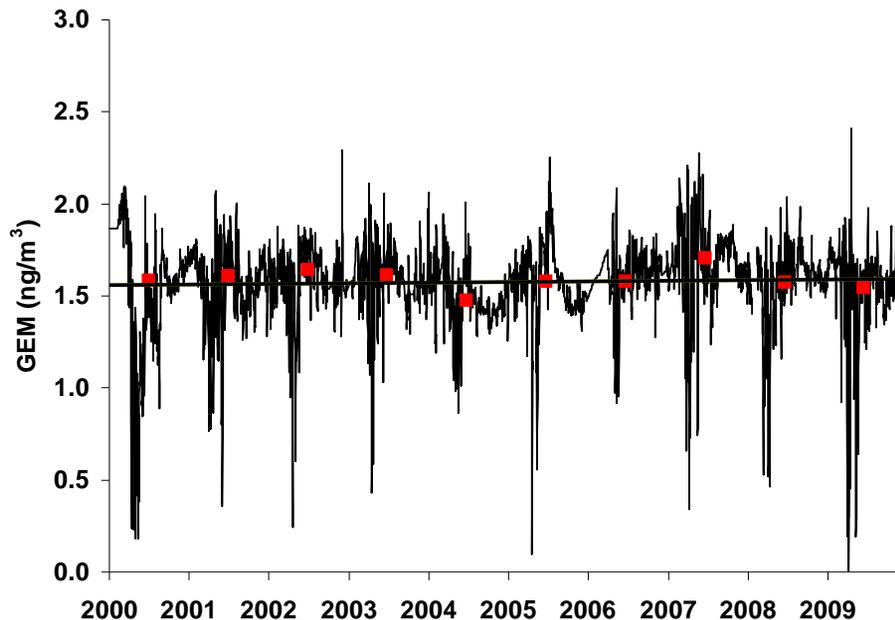


Fig. 3. The time series for GEM daily average levels including the trendline. The trendline is based on the overall slope from the trend analysis with the midpoint fixed to the median concentrations for the decade. Red squares are annual medians.

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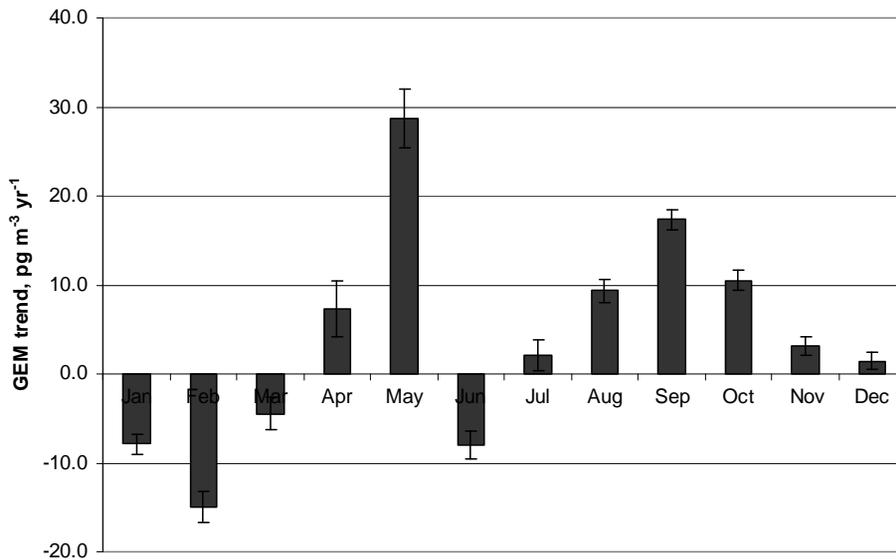


Fig. 4. Seasonal (monthly) trends in GEM base on daily average measurements 2000–2009. Error bars represent 95 % confidence limits.

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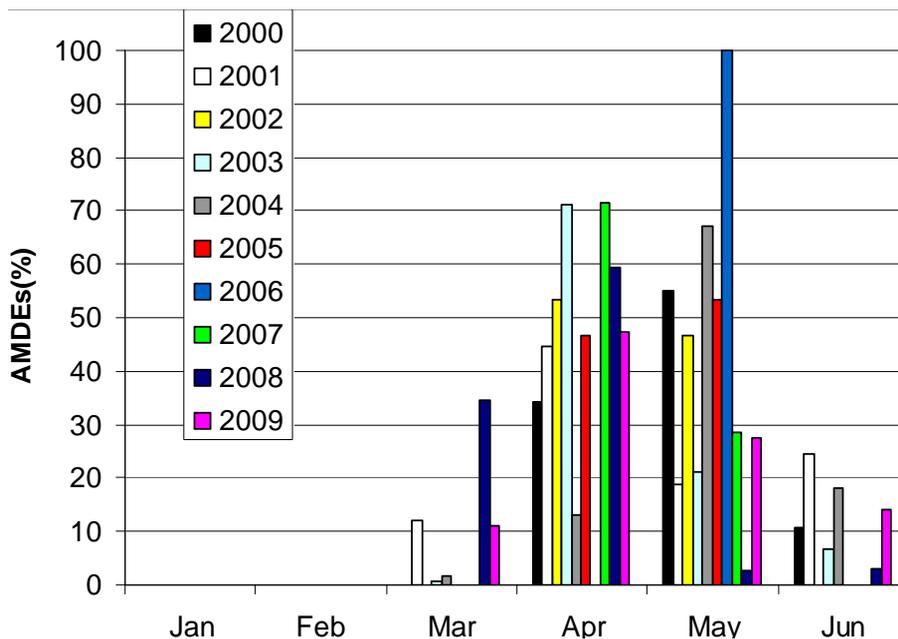
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[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)**Fig. 5a.** Distribution (%) of AMDEs at Zeppelin.

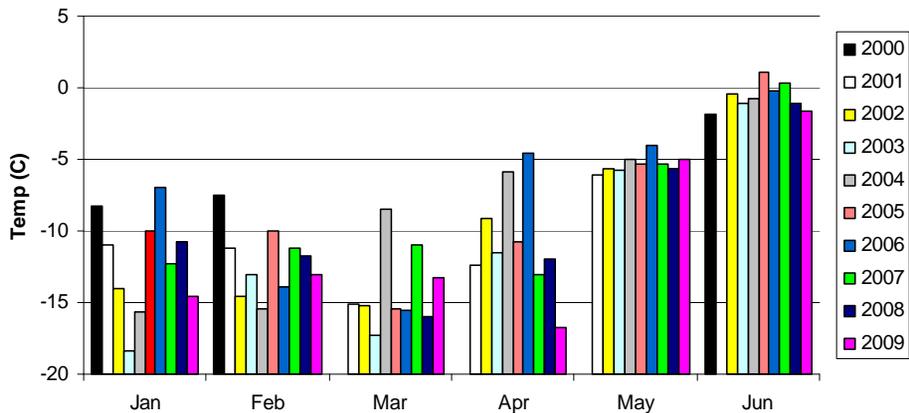


Fig. 5b. Monthly temperatures averages (January–June) at Zeppelin for 2000–2009.

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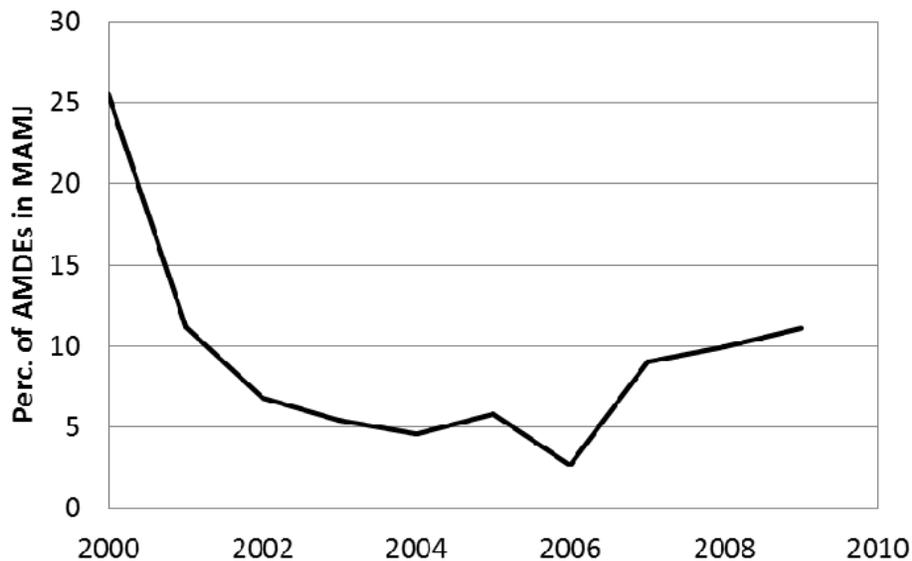


Fig. 6. Percent of AMDEs in March, April, May, June relative to the total measurements for the same period for the period 2000–2009.

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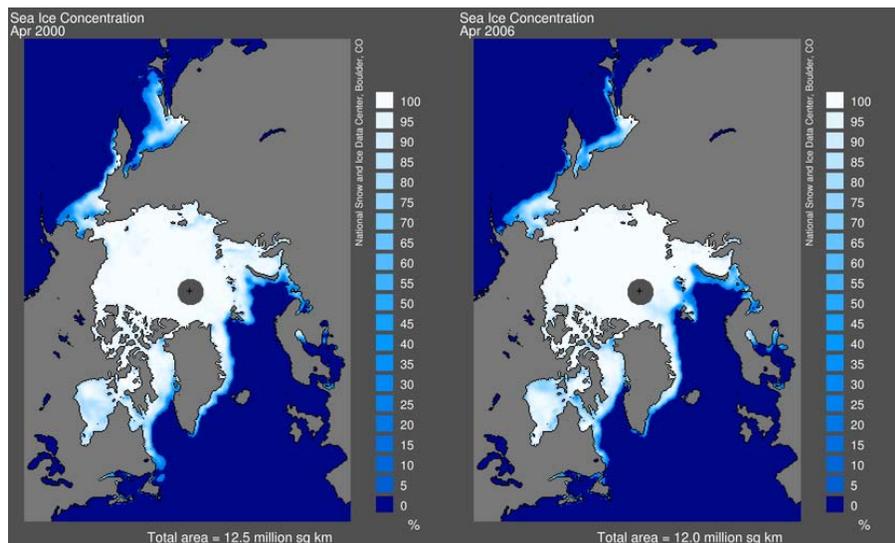


Fig. 7. Sea ice concentration maps for April 2000 and April 2006.

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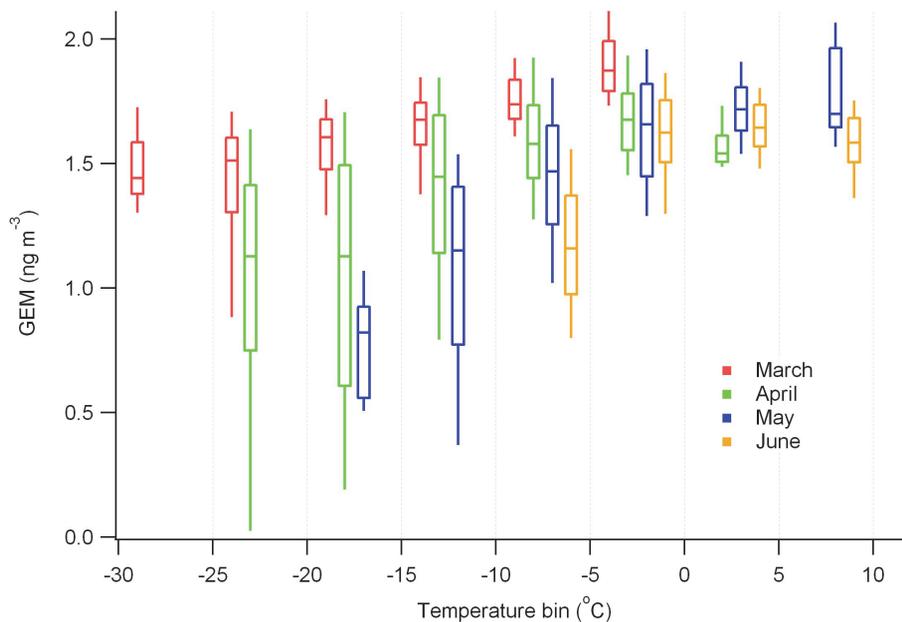


Fig. 8a. Box-and-whisker plots showing the median (horizontal line), 25th and 75th percentiles (box), and 10th and 90th percentiles (whiskers) for the distribution of GEM values in each temperature bin during each of the four AMDE months. A minimum of 20 points was required in each bin.

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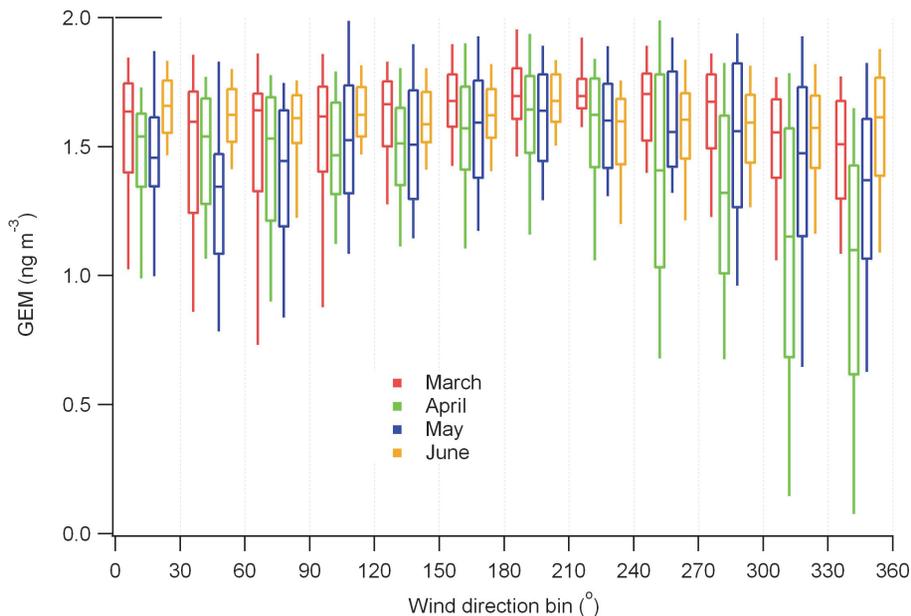


Fig. 8b. Box-and-whisker plots showing the median (horizontal line), 25th and 75th percentiles (box), and 10th and 90th percentiles (whiskers) for the distribution of GEM values in each wind direction bin during each of the four AMDE months. A minimum of 20 points was required in each bin.

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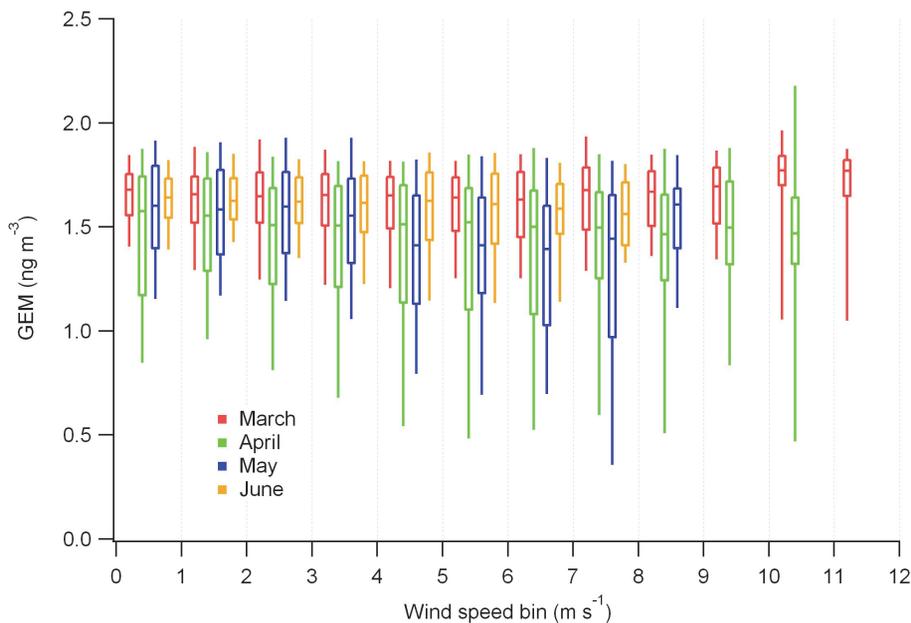


Fig. 8c. Box-and-whisker plots showing the median (horizontal line), 25th and 75th percentiles (box), and 10th and 90th percentiles (whiskers) for the distribution of GEM values in each wind speed bin during each of the four AMDE months. A minimum of 20 points was required in each bin.

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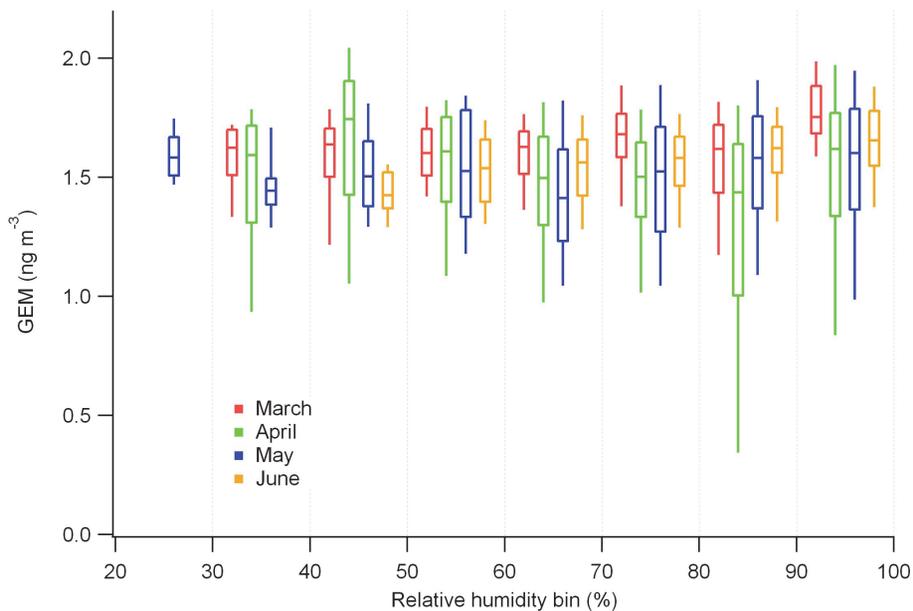


Fig. 8d. Box-and-whisker plots showing the median (horizontal line), 25th and 75th percentiles (box), and 10th and 90th percentiles (whiskers) for the distribution of GEM values in each relative humidity bin during each of the four AMDE months. A minimum of 20 points was required in each bin.

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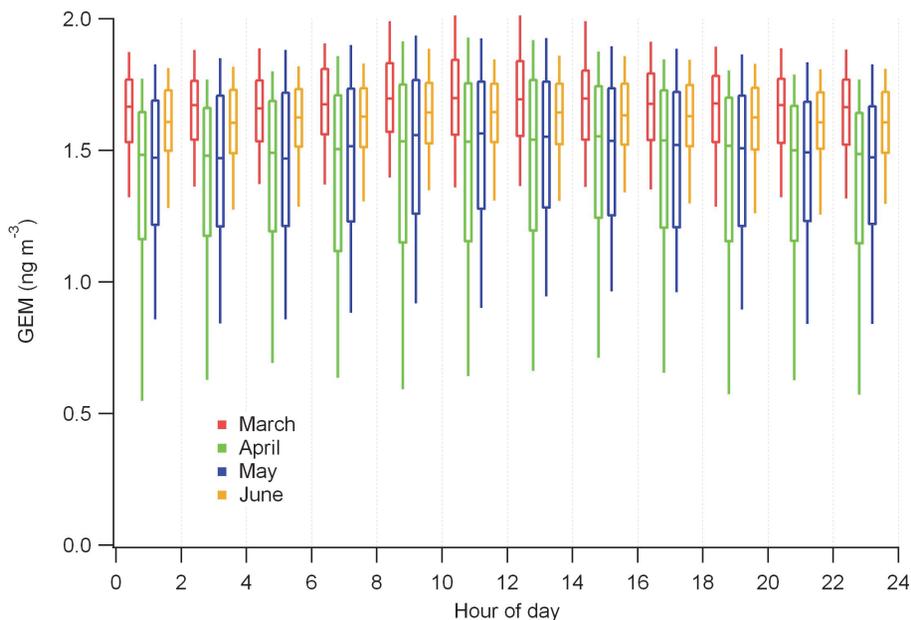


Fig. 8e. Box-and-whisker plots showing the median (horizontal line), 25th and 75th percentiles (box), and 10th and 90th percentiles (whiskers) for the distribution of GEM values in each hour of day bin during each of the four AMDE months. A minimum of 20 points was required in each bin.

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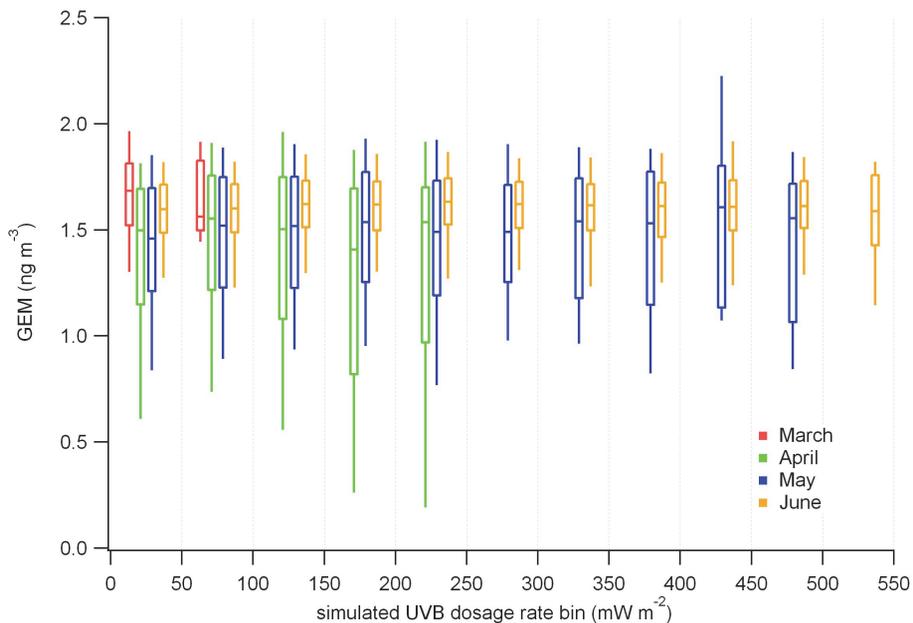


Fig. 8f. Box-and-whisker plots showing the median (horizontal line), 25th and 75th percentiles (box), and 10th and 90th percentiles (whiskers) for the distribution of GEM values in each simulated UVB bin during each of the four AMDE months. A minimum of 20 points was required in each bin.

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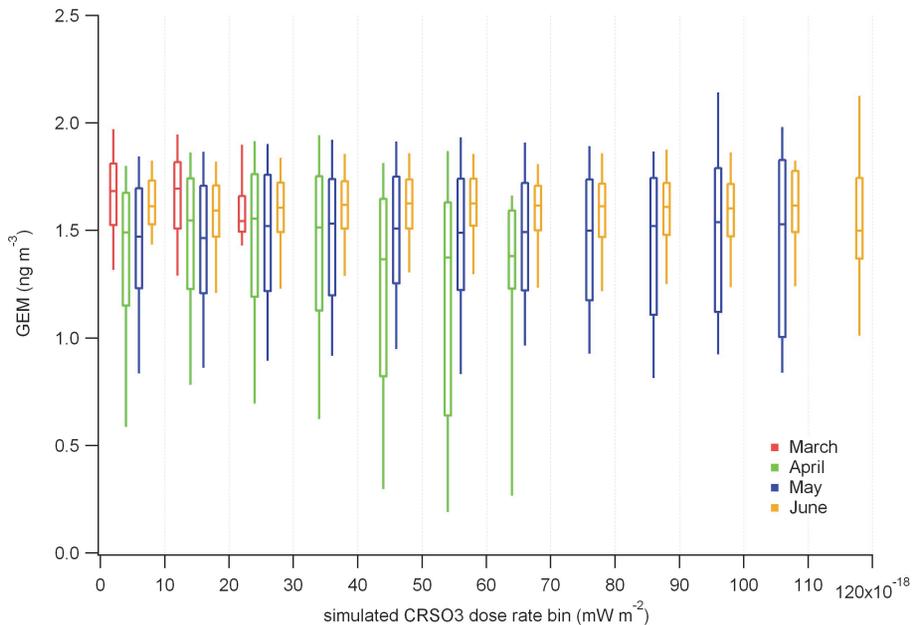


Fig. 8g. Box-and-whisker plots showing the median (horizontal line), 25th and 75th percentiles (box), and 10th and 90th percentiles (whiskers) for the distribution of GEM values in each simulated CRSO3 dose rate bin during each of the four AMDE months. A minimum of 20 points was required in each bin.

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